

Research Article

The fate of inorganic colloidal particles in Lake Brienz

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Abstract. The fate of colloidal particles in Lake Brienz and its two main tributaries, the Aare and Lütschine Rivers, was studied over a period of more than a year. Lake colloid loads from the unmodified Lütschine fluctuated significantly across seasons. The highest loads are in summer due to glacier melting. Colloid loads from the Aare reflected the seasonal water use by upstream hydropower plants. A significant number of mineral particles, even outside the theoretical non-settling range, remain in suspension in the epilimnion of Lake Brienz in summer because of the mineralogical nature and shape of the particles and the high input loads. Only 1730 of the 16,800 tons of colloidal particles that entered the lake during the period from 1 June 2004–31 July 2005 were exported through the lake outlet. The remainder was presumably lost in the

lake through coagulation-sedimentation processes. Coagulation properties of non-fractionated colloid samples in quiescent conditions were studied in the laboratory both in the absence and presence of different organic compounds representative of natural organic matter. Lake Brienz colloids coagulate slowly, as expected from particles (mainly phyllosilicates) bearing a net negative surface charge. Slightly higher coagulation rates were recorded in March and April during the spring algal bloom, which suggests that colloid coagulation is enhanced by carbohydrates. This effect was confirmed in laboratory experiments. However, the concentration of natural organic matter in Lake Brienz is so low that it is not having any significant effect on the fate of inorganic colloids.

Key words. Lakes; rivers; particle size distributions; coagulation; sedimentation; SPC; turbidity.

Introduction

Lakes that drain glacier-melt waters have high turbidities due to the suspension of inorganic particles from glacier abrasion (Koenings et al., 1990). As a result of the high concentration of suspended particles, which backscatter a portion of the incident solar radiation (Hecky, 1984), these lakes are characterised by a smaller trophogenic zone and cooler summer water temperatures (Koenings et al., 1990) compared to

non-glacial lakes. High mineral turbidity modifies food webs through direct and indirect effects at all trophic levels (Hart, 1988; Cuker et al., 1990; Rellstab et al., 2007). Although the extent suspended clays influence lake trophic organisation depends on mass loading rates and mineral particle size and mineralogy (Threlkeld and Søballe, 1988; Cuker et al., 1990; Cuker and Hudson, 1992), this issue has been little studied, and the influence of particles in the colloidal size have never been investigated. As they are small in size, colloids have long settling times in water, typically from days to months. The long residence times of colloidal particles, together with their highly

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specific surfaces, makes it likely that significantly influence lake optical properties (Jassby et al., 1999).

Within the framework of an extensive research project into the causes and consequences of the current oligotrophic status of Lake Brienz, Switzerland (Wüest et al., 2007), the fate of mineral colloid particles was studied in the lake and its tributaries. Quantitative studies of colloid behaviour at the scale of a whole water body (e.g. lake) are rare because of the lack of adequate measuring techniques. Most existing studies either give only qualitative information or are devoted to a specific aspect of colloid behaviour. The methodology used in this study included different complementary techniques in the field and in the laboratory (Fig. 1). Since coagulation plays a vital role in colloid removal in natural systems, field measurements have been supplemented by laboratory measurements. To test the expectation that an increase in lake productivity would favour colloid coagulation and particle removal, experiments were performed in the presence of different types of natural organic matter (NOM). Since low productivity (Finger et al., 2007), light availability (Jaun et al., 2007) and coagulation processes are tightly interrelated, special attention was paid to epilimnion waters because of their particular importance in terms of water clarity and productivity. The methods section includes a detailed discussion on the capabilities of the different techniques and calculation methods used. The term 'colloid' has been used with different meanings by different authors (Filella, 2007). In the following text, the term 'colloid' is used to refer to entities between 0.1 and 2 μm (analytical window of the Single Particle Counter (SPC) used) and, unless otherwise stated, the term 'particle' is only applied for those $> 2 \mu\text{m}$.

Study systems

Lake Brienz is in Switzerland at an altitude of 564 m asl. It has a surface area of 29.8 km² and a volume of 5.1 km³, and is a typical peri-alpine lake. The lake is deep (maximum depth: 260 m), rectangular in shape (~14 km x 2 km), and has an average hydraulic residence time of 2.7 y. It almost was mesotrophic in the late 1970 s and early 1980 s but now it is considered ultra-oligotrophic (Müller et al., 2007). Lake Brienz has two main tributaries: the River Aare and the River Lütschine. Both are glacial rivers and drain catchment areas of 554 km² and 379 km², respectively, of which 21 % and 17 %, respectively, are covered by glaciers. In contrast to the Lütschine, the hydraulic regime of the Aare was significantly modified during the 20th century by an extensive damming system used for

production of electricity. A more detailed description of Lake Brienz and its tributaries is available in Wüest et al. (2007).

Methods

Between June 2004 and October 2005, eight field campaigns were carried out (9–12 June 2004, 30 August–2 September 2004, 17–20 November 2004, 20–23 February 2005, 21–24 March 2005, 25–29 April 2005, 19–23 July 2005 and 3–7 October 2005). Samples were collected in the Lütschine (46°38'52"N, 7°52'29"E), Aare (46°44'37"N, 8°3'2"E), and in the centre of Lake Brienz (46°43'2"N, 7°56'59"E). Lake water samples were taken using a membrane pump, with the end of the tube directly attached to a multi-parameter probe (Züllig HTP-D, Switzerland). Samples for additional coagulation experiments were taken in August and September 2005 from the shore of Lake Brienz at Iseltwald (46°42'46"N, 7°57'46"E), Lake Thun at Krattigen (46°40'15"N, 7°42'59"E) and at an outlet of the Grimsel Kraftwerke Oberhasli (KWO) hydropower plant at Innertkirchen (46°42'29"N, 8°13'36"E). River samples were collected in bottles at a depth of about 10 cm. The conductivity and temperature of the river water were recorded. Samples for colloid analysis and coagulation experiments were collected in thermostated bottles (1 or 2 L) and immediately transported to the *on site* laboratory. Samples for total suspended solids were collected in polyethylene bottles (2 L).

Single Particle Counter

Number particle size distributions (PSD) were measured with a Single Particle Counter (SPC) in the range 0.1 to 2 μm using unfiltered samples. The instrument (Particle Measuring Systems, Boulder, CO, USA) is based on the principle of light scattering by single particles. The measuring procedure is detailed in Rossé and Loizeau (2003). Analyses were always performed *on site* within 1 h of sample collection. The physical meaning of the SPC-measured sizes for non-spherical particles is not currently known. Non-imaging methods, such as light scattering techniques, yield equivalent spherical diameters. This is the diameter of a sphere that would give the same signal as the actual particle. Different techniques may yield different equivalent spherical diameters for the same particle. With the SPC, the light scattered by a single particle is collected and the light intensity integrated between 50 and 130° (Knollenberg and Veal, 1991). The principle is relatively similar to the small angle, static light scattering (SLS) technique where the intensity of scattered light is measured over a range of forward

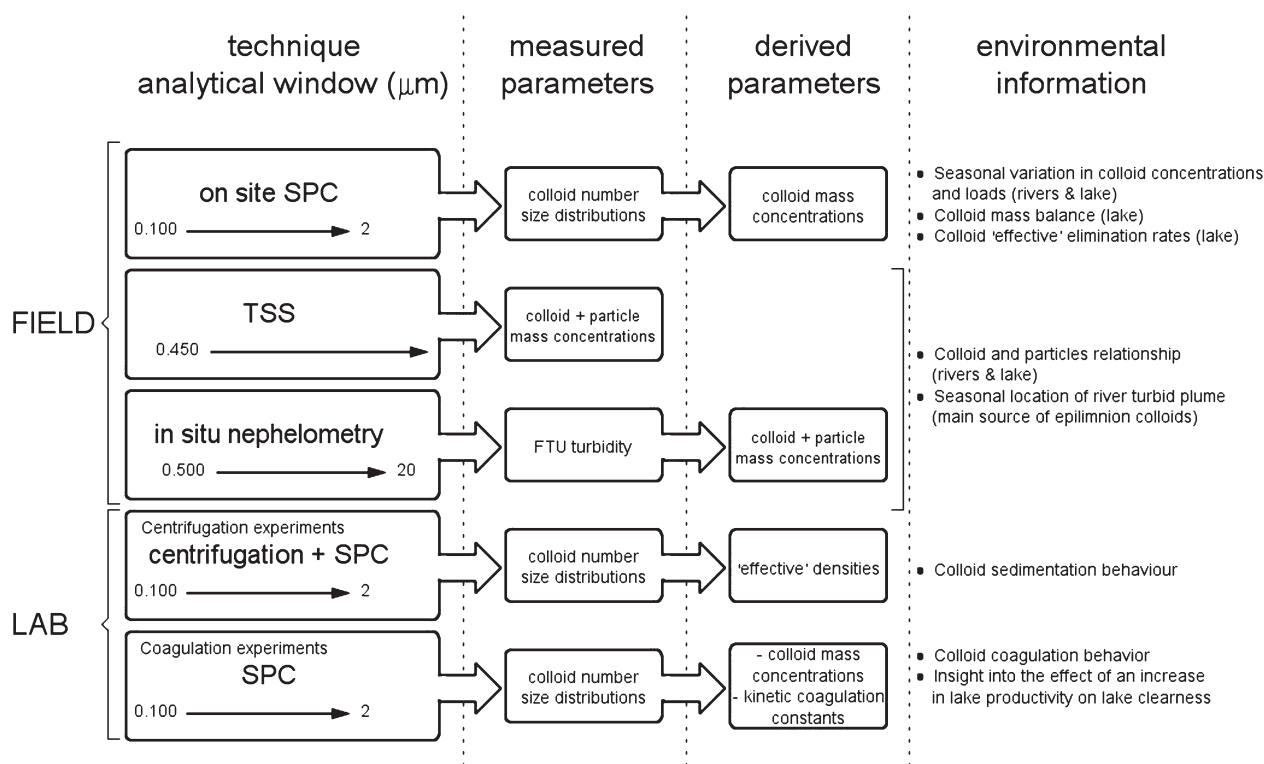


Fig. 1. Methodological approach used in this study.

angles. SLS measurements are often evaluated as radii of gyration (Schurtenberger and Newman, 1993; Sun, 2004). Since the SPC is calibrated by measuring the radii of spherical latex beads, the SPC-measured radii for natural pseudo-spherical particles should be close to the particle geometric radii. In the case of plate-like particles, the radii of gyration are probably the best approximation of the SPC-measured radii.

Total suspended solids

Total suspended solid (TSS) concentrations were determined by filtering a known volume of water (between 200 mL and 2 L, depending on suspended matter content). Filters (cellulose nitrate, 0.45 μm , 50 mm, Schleicher et Schuell) were rinsed with 1 L of Milli-Q water, dried (3 h at 60 °C) and weighed. Once the sample had been filtered, the filters were dried (3 h at 60 °C) and weighed again.

Turbidity

Turbidity, temperature and conductivity profiles were measured in Lake Brienz with a multi-parameter probe (Zülig HPT-D, Switzerland). Turbidity measurements were taken using a light scattering nephelometer that works on the basis of forward scattering of light by suspended particles. Calibration was performed with a formazine standard and results are expressed in formazine turbidity units (FTU). As

turbidity is a function of the size and nature of the particles present, it is difficult to ascertain which particles (type and size) are responsible for the measured turbidity in a particular system. Since the nephelometer used (630 nm wavelength, maximum scattering angle of 6°) measures suspended particles < 20 μm (Morel, 1973), and it is well known that scattering efficiency rapidly reduces with decreasing particle size < 1 μm (Mishchenko et al., 2002), the turbidity recorded is probably caused by inorganic particles < 20 μm with colloidal particles < 0.5 μm contributing little or none. Since NOM concentrations are extremely low in Lake Brienz (Chanudet and Filella, 2007), turbidity is due almost exclusively to the presence of mineral particles.

Calculation of particle concentrations from turbidity data (Hoffman and Dominik, 1995; Finger et al., 2006) is complex because the relationship between turbidity and TSS not only depends on the type of particles present but also on the different measuring windows associated with both parameters. Three statistically different turbidity-TSS correlations were obtained from three different campaigns in Lake Brienz (Table 1). The results show that two waters containing similar (in number and type) particles < 20 μm may give similar turbidity values while having very different TSS concentrations when the concentration of particles > 20 μm differs.

Table 1. Total suspended solids (TSS), turbidity, particle total mass, total colloid number and mass concentrations for Lake Brienz and its tributaries.

System	Date	TSS ^b (mg L ⁻¹)	Turbidity ^c (FTU)	Total mass ^d (mg L ⁻¹)	Total number 0.1–2 µm ^b (×10 ⁷ mL ⁻¹)	Total mass 0.1–2 µm ^c (mg L ⁻¹)	Total mass 0.45–2 µm ^c (mg L ⁻¹)	% colloid : total mass
Lake Brienz, 1 m depth	Jun 2004		1.8 ± 0.2	1.7 ± 0.7	3.61 ± 0.07	0.38 ± 0.01	0.155 ± 0.001	22
	Sep 2004		5.6 ± 0.2	5.6 ± 0.2	4.86 ± 0.13	0.76 ± 0.04	0.41 ± 0.01	13
	Nov 2004		2.5 ± 0.4	2.2 ± 0.8	2.63 ± 0.04	0.41 ± 0.01	0.217 ± 0.002	18
	Feb 2005		1.03 ± 0.02	1.2 ± 0.6	1.73 ± 0.03	0.28 ± 0.01	0.145 ± 0.003	23
	Mar 2005		0.72 ± 0.07	1.1 ± 0.6	1.76 ± 0.05	0.23 ± 0.01	0.102 ± 0.004	21
	Apr 2005		1.52 ± 0.01	1.6 ± 0.6	2.32 ± 0.03	0.38 ± 0.01	0.223 ± 0.004	24
	Jul 2005	4.7 ± 0.1	6.5 ± 0.1	4.7 ± 0.7	6.55 ± 0.03	1.20 ± 0.02	0.71 ± 0.02	26
	Aug 2005				6.35 ± 0.03	0.92 ± 0.02	0.55 ± 0.01	
	Sep 2005				5.96 ± 0.04	0.93 ± 0.02	0.56 ± 0.01	
	Oct 2005	3.5 ± 0.1	4.18 ± 0.03	3.2 ± 0.6	5.53 ± 0.05	0.85 ± 0.01	0.476 ± 0.004	27
Lake Brienz, turbidity peak ^a	Jun 2004		12.3 ± 0.4	8.4 ± 0.8	2.92 ± 0.05	0.93 ± 0.01	0.70 ± 0.01	11
	Sep 2004		10.7 ± 0.6	10.6 ± 0.6	6.22 ± 0.10	1.50 ± 0.06	1.05 ± 0.05	14
	Jul 2005	7.5 ± 0.1	10.6 ± 0.1	7.3 ± 0.7	13.3 ± 0.1	2.11 ± 0.04	1.21 ± 0.03	29
	Oct 2005	7.7 ± 0.1	8.9 ± 0.1	6.2 ± 0.7	10.3 ± 0.2	1.82 ± 0.02	1.10 ± 0.01	29
River Aare	Jul 2005	44.0 ± 0.4			68.9 ± 0.5	8.81 ± 0.10	4.59 ± 0.06	20
	Oct 2005	33.2 ± 0.3			43.2 ± 0.3	5.66 ± 0.07	2.87 ± 0.05	17
River Lütschine	Jul 2005	44.0 ± 0.4			36.0 ± 0.1	6.65 ± 0.06	3.19 ± 0.03	15
	Oct 2005	39.0 ± 0.2			21.3 ± 0.3	4.13 ± 0.04	2.53 ± 0.02	11

^a No turbidity peaks were present from November 2004 to April 2005.

^b Error: 1 SD.

^c Error (1 SD) calculated from a 40 cm water layer centred on the stated depth.

^d Calculated from the turbidity-TSS calibration performed in July 2005 (turbidity = 1.57 TSS – 0.92; $r^2 = 1.00$) except for September 2004 (turbidity = 1.02 TSS – 0.06; $r^2 = 0.94$) and October 2005 (turbidity = 0.92 TSS + 1.14; $r^2 = 0.68$) where calibration performed during these campaigns was used.

^e Calculated by assuming a density of 2.65 g mL⁻¹. Error: 1 SD.

Estimation of colloid mass concentrations from number particle size distributions

Mass-based PSD can be calculated from number PSD by multiplying the mean mass in each size class (estimated by taking the diameter and a mean density) by the number of particles. In practice, the procedure is complex because of uncertainties associated with the physical meaning of sizes measured by SPC and with the density assigned to the particles. The main inorganic colloids (biotite, chlorite, illite, feldspars and quartz) present in Lake Brienz and its tributaries (Chanudet and Filella, 2006; Filella and Chanudet, 2006) have a relatively similar plate-like shape and specific densities near 2.65 g mL⁻¹ (Klein, 2002). If the radii given by the SPC are close to the radii of gyration for this type of particle, the actual colloid volumes and the volumes corresponding to spherical colloids with the SPC-measured radius will be close and thus 2.65 g mL⁻¹ can be used as the density associated with the measured radii. Any errors associated with the number to mass transformation procedure, other than random errors in measurements, should be seen as systematic errors and, as such, their effect cannot be expressed with standard deviations.

One-box mass balance calculations

One-box mass balance calculations were performed for the period 1 June 2004 to 31 July 2005. River loads

were calculated with the program FLUX (1999) from concentration data and continuous flow records. FLUX has five possible methods of estimating loads. The most appropriate method is selected by the user according to the quality of the log(concentration) versus log(flow) correlation (Walker, 1999). A method that applies the concentration/flow regression equation individually to each daily flow value is preferable when the correlation is good, as in the case of the Aare and Lütschine in this study. Daily flow values were provided by the Federal Office for the Environment of Switzerland (FOEN, 2005). The errors in flows were normally between 2 and 5% (and up to 10% during floods) (FOEN, 2005) and those in colloid concentrations were below 5% (Table 1). The error in calculated loads was estimated by using the jackknife procedure.

‘Effective’ lake colloid elimination rates

‘Effective’ lake colloid elimination rates were estimated in the epilimnion by applying a simple one-box budget calculation method that takes into account particle elimination. At time t , the mean colloid concentration, m (g L⁻¹), is calculated by considering two contributions: (i) the mean colloid concentration remaining from the previous time step in the integration procedure assuming that colloid elimination follows second order rate kinetics (see below) (first

term in eq. 1), and (ii) the difference between riverine input and output at time t (second term in eq. 1)

$$\frac{dm}{dt} = -k[m(t)]^2 + \frac{I(t) - O(t)}{V(t)} \quad (1)$$

where: t = time (h), $m(t)$ = mean colloid concentration in the epilimnion at time t (g L^{-1}), k = colloid elimination rate constant ($\text{L g}^{-1} \text{h}^{-1}$), $I(t)$ = colloid input (rivers) at time t (g h^{-1}), $O(t)$ = colloid output (rivers) at time t (g h^{-1}), $V(t)$ = 'effective' lake volume at time t (L). $I(t)$ and $O(t)$ were calculated from SPC measurements with the program FLUX (1999). The initial colloid concentration used in the calculations is the one measured in the first sampling campaign (June 2004). Given the seasonal differences in lake water, two different 'effective' lake volumes were considered in the calculations: the whole lake depth (260 m) was taken from 1 November 2004 to 30 April 2005 (homogeneous colloid concentration in the whole water column) and the volume corresponding to a depth of 40 m (epilimnion) for the rest of the study period.

Laboratory experiments

Sedimentation. 'Effective' particle densities were estimated by combining SPC measurements and centrifugation. Colloids and particles were partially removed from water by centrifugation and those remaining in suspension were measured by SPC. From the classical Stokes' law as applied to centrifuged spherical colloids (Lienemann et al., 1998), the theoretical fraction of remaining colloids and particles after centrifugation is given by

$$\% \text{ remaining colloids \& particles} = 100 \left[1 - \frac{x_2 \left(1 - \exp \left(-\frac{\omega^2 \phi^2 (\rho - \rho_0) t}{18 \eta} \right) \right)}{x_2 - x_1} \right] \quad (2)$$

where: ω = centrifuge rotation speed (radian s^{-1}), ϕ = particle diameter (cm), ρ = particle density (g mL^{-1}), ρ_0 = water density (1 g mL^{-1}), t = time (s), η = water viscosity ($\text{g cm}^{-1} \text{s}^{-1}$), x_1 and x_2 = minimum and maximum distances in the tube relative to the axis of the centrifuge (cm). The percentage of remaining particles was calculated from equation (2) for several time steps by adjusting the particle density to fit the measured data.

Coagulation. Coagulation experiments were designed to determine the rate that colloids with sizes in the SPC range coagulate under quiescent-settling conditions. After sampling, water was left undisturbed in thermostated bottles (2 L) for the full duration of the

coagulation experiment. No homogenization was carried out or agitation applied before measurements were taken. The sample was pumped directly from the thermostated bottles 4 cm below the water surface and changes in number concentration and PSD over time were measured by SPC at several points between 1 h and ~ 4 weeks after sampling. The experimental design and the measuring method used will reflect not only coagulation, but any process leading to changes in the amount and size of colloids present in the size range 100 – 2000 nm. In the following section it is assumed that coagulation is the main process responsible for the changes observed.

Coagulation in the presence of model NOM. Coagulation was studied after the addition of selected organic compounds to lake water. Measurements were taken on three occasions (August, September and October 2005). The organic substances added were galacturonic acid, alginic acid, dextran, polygalacturonic acid, and IHSS Suwannee River fulvic acid (see Table 3 for concentrations). Once the appropriate amount of each organic compound added to 1 L of unfiltered water, samples in thermostated bottles (5 x 1 L per experiment) were placed onto a horizontal rotating system with the axis of rotation parallel to that of the bottles. Bottles could not turn on their own axis. A system with a very low rotation speed (2 rpm) was used to ensure that the suspension remained homogeneous throughout the experiment while avoiding coagulation induced by a high velocity gradient. PSD measurements were taken over 28 days in August and 12 days in September and October 2005. Two hundred mL aliquots were taken from the bottles at different times for PSD measurements. The bottles were immediately returned to the rotating system after sampling. No bacterial growth was observed by TEM in the absence or presence of any type of NOM within 2 weeks.

Transmission Electronic Microscopy. Transmission Electronic Microscopy (TEM) was performed with a Philips CM12 microscope (source W, 80 kV accelerating voltage) coupled with a CCD camera. Specimens were prepared by centrifugation (1700 g, 90 min, Roto Silenta centrifuge, Hettish, CT, USA) in conventional tubes with specimen grids (200 mesh, carbon coated, collodion covered; Plano GmbH, Germany) at the bottom. Considering a spherical shape and a density of 1.6 g mL^{-1} , the minimum size of colloids quantitatively collected onto TEM grids is about 150 nm.

'Laboratory' related calculations

Quantification of the coagulation properties of natural colloids from PSD data is complex because of the

heterodispersive character of such particles. In the SPC size range, where colloidal Brownian motion is the main physical interparticle contact mechanism, the change in the total number concentration of particles N (L^{-1}) of a monodispersive colloidal suspension in the early stage of coagulation can be described by

$$\frac{dN}{dt} = -k_n N^2 \quad (3)$$

where: t = elapsed time (h), k_n = coagulation rate constant ($L h^{-1}$). Although equation (3) is based on the assumption of a constant, non size-dependent collision kernel and is therefore applicable only to the early stages of coagulation of initially monodispersive particles, it has proven to correspond reasonably well to numerical calculations for predicting the number decay even for late stages (Hidy, 1965) and has been used for polydisperse aerosols (Lee, 1983) and to describe the aggregation rates of natural particles such as lake iron oxides (initial particles in the range $0.05 - 0.5 \mu m$) (Tipping and Ohnstad, 1984). The application of equation (3) to heterodispersive samples can be justified on theoretical grounds if Swift and Friedlander's (1964) self-preserving hypothesis holds throughout the coagulation.

Results

Field observations

Colloid seasonal variations in the lake and the tributary rivers. Seasonal colloid PSD were measured in Lake Brienz and its tributaries by SPC. The number concentration of colloidal particles was found to increase with decreasing size in all samples. Mean colloid number and mass concentrations calculated from SPC data are shown in Table 1. The seasonal variation in mass colloid concentrations and loads in the Aare and Lütschine is shown in Figures 2 and 3, respectively. The values given in Figure 3 for the period August-September 2005 probably grossly underestimate the number of colloids that actually entered the lake because they fail to account for the flood of August 2005. This flood brought a significant, but unknown, amount of colloids into Lake Brienz, not only via the Aare and Lütschine but also directly through landslides and overflowing smaller tributaries. The values shown in Figure 3 should be taken as conservative estimates since they include the effect of the Aare and Lütschine high flows, but not of any other input of colloids or of the probable increase in colloid concentrations in both rivers as a result of flooding.

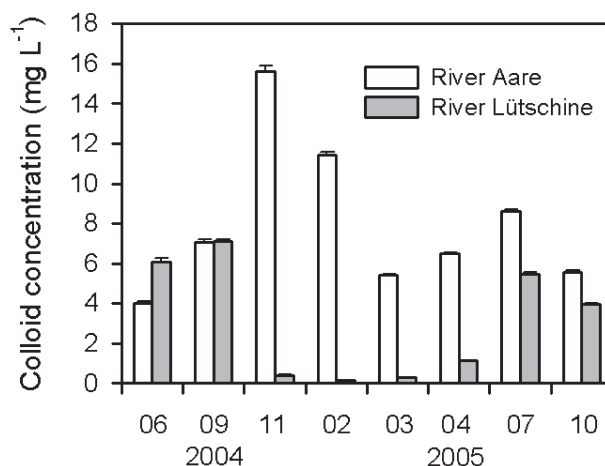


Fig. 2. Colloid concentrations in the Aare and Lütschine from June 2004 to October 2005.

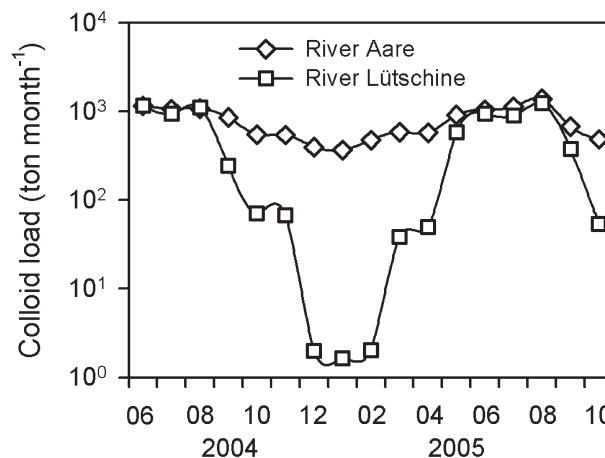


Fig. 3. Colloid input loads in Lake Brienz from the Aare and Lütschine from June 2004 to October 2005.

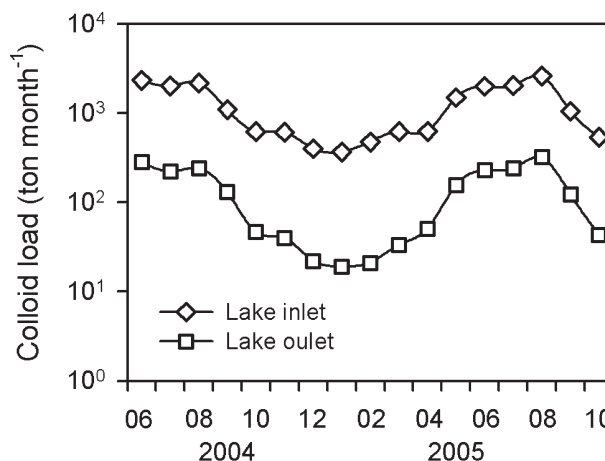


Fig. 4. Comparison of colloid input and output loads in Lake Brienz from June 2004 to October 2005.

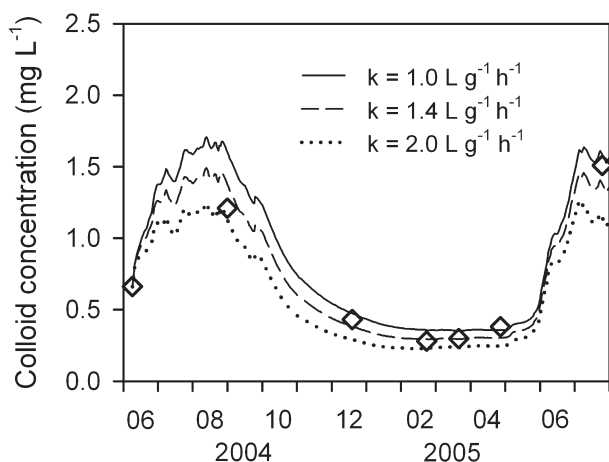


Fig. 5. Effect of the variation in lake 'effective' colloid elimination rate constant, k , ($\text{L g}^{-1} \text{h}^{-1}$) on the mean colloid concentration in Lake Brienz. Measured concentrations: \diamond .

the Lütshine, respectively) and the colloid flux leaving the lake was 1730 ± 414 tons.

'Effective' lake colloid elimination rates. The elimination of colloids observed in the lake was estimated through the calculation of an 'effective' colloid elimination rate constant. The effect of the variation of the 'effective' colloid elimination rate, k , on the calculated colloid concentrations is shown in Figure 5. Measured concentrations in the epilimnion for the different field campaigns are also shown. For a k value of $1.4 \text{ L g}^{-1} \text{h}^{-1}$ (least square method), the model explains the measured data well ($r^2 = 0.97$). Assuming a mean colloid concentration of 0.69 mg L^{-1} , a k value of $1.4 \text{ L g}^{-1} \text{h}^{-1}$ gives a mean colloid half life time of about 43 days.

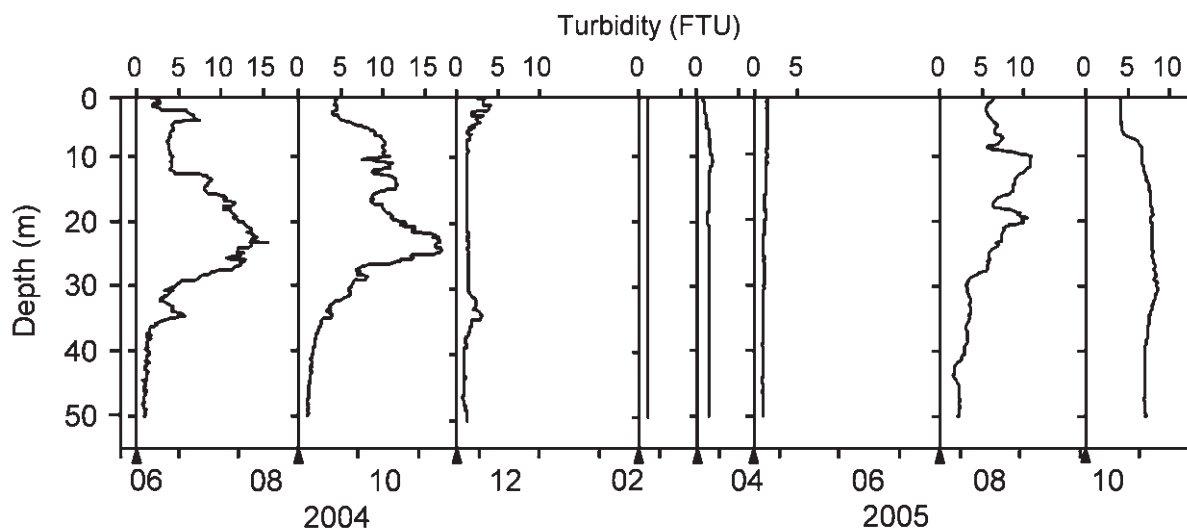


Fig. 6. Turbidity (FTU) profiles in Lake Brienz for the eight sampling campaigns.

Colloid mass balance in Lake Brienz. A simple one-box model was applied to the assessment of colloids in Lake Brienz, assuming that colloids only enter the lake through the Aare and Lütshine and exit through its outlet. Budget calculations were done only from 1 June 2004 to 31 July 2005 to exclude the flood of August 2005. Total colloid masses in the epilimnion were 1030 and 1100 tons in June 2004 and July 2005, respectively. Thus, colloids can be considered to be in steady state on an annual scale. The input and output of colloids in Lake Brienz between June 2004 and October 2005 is shown in Figure 4. The variation in input and output loads is similar, the colloid flux at the outlet of the lake being about $10 \times$ lower than the inputs. Between 1 June 2004 and 31 July 2005, the colloid flux entering the lake was $16,800 \pm 2410$ tons ($10,700 \pm 1710$ and 6100 ± 1700 tons for the Aare and

Relationship between colloids and particles. Turbidity profiles measured during the field campaigns are shown in Figure 6. In winter, turbidity was low and homogeneous throughout the lake water column. For example, turbidity values ranged between 1.0 and 1.1 FTU in the uppermost 100 m in February 2005. In summer, a strong turbid layer appeared above 40 m and extremely high turbidity values were recorded (e.g. up to 17 FTU were measured at a depth of 23 m in September 2004). Light transmission data for 2003 and 2004 also confirm the existence of this layer (GBL, 2005) in previous years. This turbid layer remains above the thermocline throughout summer, reflecting river intrusion.

Rapidly changing turbidity values in profiles recorded successively showed the dynamic nature of the turbid plume in the lake. Figure 7a shows turbidity

profiles recorded at the same point (centre of the lake, equidistant from both rivers) over two hours. These profiles show a marked difference in the position of the turbidity peaks. The spatial variability of the turbidity profiles in the centre of the lake and in the two river mouths is shown in Figures 7b–d. Both the shape (depths of the different peaks) and the absolute turbidity values vary significantly between two points about 500 m apart (see Fig. 7e for the location of sampling points). This level of variability must be taken into account when considering any turbidity profile measured in Lake Brienz as well as the validity of any derived parameter (i.e., particle mass concentrations).

Turbidity-TSS correlations have been used to estimate particle concentrations from turbidity data (Table 1). Colloid mass concentrations account for 22% of the TSS at a depth of 1 m and 21% at the turbidity peak in Lake Brienz, and for a somewhat lower proportion in the two rivers (~16%). Thus, about 80% of the suspended particles are $> 2 \mu\text{m}$.

Laboratory results

Sedimenting behaviour of Lake Brienz colloids and particles. The sedimenting behaviour of Lake Brienz colloids and particles was studied by determining ‘effective’ densities in the laboratory. The results (Fig. 8) show a decrease in the ‘effective’ density with size in the colloidal size range: larger colloids behave as if they have a lower density than smaller ones. The results suggest that Lake Brienz colloids and particles sediment more slowly than Stokes’ law predicts. The difference becomes even more pronounced as the size of the colloids and particles increases. It has often been observed that natural colloids form loose aggregates characterized by low sedimenting velocities. This is not the case for Lake Brienz where TEM observations have shown the presence of mainly ‘large’ individual colloids and particles and of relatively few, compact aggregates.

Coagulation behaviour of Lake Brienz colloids. The number- and mass-colloid concentration changes with time for Lake Brienz (1 m depth) and for the Aare and Lütischine are shown in Figure 9. Plots of the inverse of the mean colloid number concentrations and of the mean colloid mass concentrations against time were linear (correlation coefficients always ≥ 0.91) for all coagulation experiments, showing that the kinetics of coagulation is adequately described by second order kinetic constants. The coagulation rate constant values are shown in Table 2.

Note that number-based and mass-based rate constants do not give equivalent information. Number and mass constants are calculated from mean number-

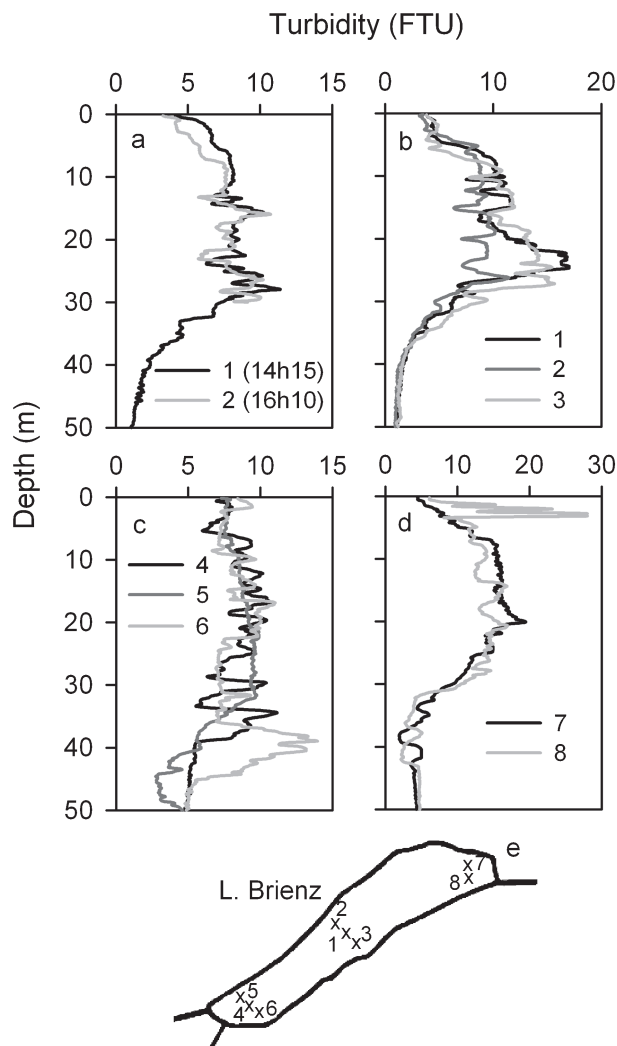


Fig. 7. Turbidity (FTU) profiles in (a) centre of Lake Brienz ($46^{\circ}43'11''\text{N}$, $7^{\circ}57'12''\text{E}$) at 14:15 and 16:10 (point 1 in figure 2e), (b) centre of Lake Brienz (point 1 in figure 2e) about 600 m NW ($46^{\circ}43'30''\text{N}$, $7^{\circ}56'59''\text{E}$) and SE ($46^{\circ}43'2''\text{N}$, $7^{\circ}57'35''\text{E}$) (points 2 and 3 in figure 2e), (c) mouth of the Lütischine ($46^{\circ}41'45''\text{N}$, $7^{\circ}54'19''\text{E}$) (point 4 in figure 2e) about 500 m NW ($46^{\circ}41'56''\text{N}$, $7^{\circ}53'58''\text{E}$) and SE ($46^{\circ}41'32''\text{N}$, $7^{\circ}54'29''\text{E}$) (points 5 and 6 in Figure 2e), (d) mouth of the Aare ($46^{\circ}44'54''\text{N}$, $8^{\circ}1'38''\text{E}$) and about 500 m S ($46^{\circ}44'39''\text{N}$, $8^{\circ}1'43''\text{E}$) (points 7 and 8 in figure 2e). Turbidity profiles shown in a and b were measured on 30 August 2004. Turbidity profiles shown in c and d were measured on 31 August 2004.

derived and mean mass-derived colloid concentrations, which are inherently weighted in a different way: while particle number concentrations are dominated by the ‘smaller’ colloids (colloids $< 0.2 \mu\text{m}$ make up about 85% of the total number of colloids between 0.1 and $2 \mu\text{m}$), mass concentrations are mostly influenced by the ‘bigger’ colloids (colloids $> 0.2 \mu\text{m}$ make up about 80% of the total colloid mass). Number-based rate constants will therefore mostly reflect the removal of ‘small’ colloids, whereas the

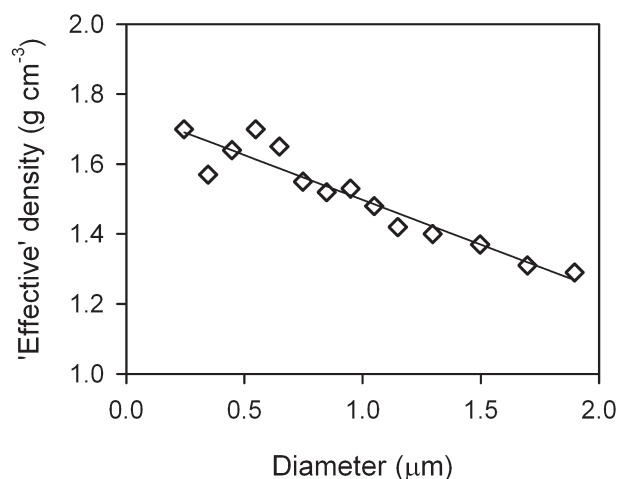


Fig. 8. 'Effective' density of Lake Brienz particles (September 2005) calculated from centrifugation experiments as explained in the text.

mass-based rate constants reflect the elimination of 'bigger' colloids.

Coagulation in the presence of organic substances. The results obtained for colloid coagulation experiments performed in the presence of organic substances, representative of different types of natural organic matter, are shown in Table 3 and Figure 10. Fulvic acids increased colloid stability. Of the carbohydrates tested, only the monosaccharide galacturonic acid had a stabilising effect. Because its charge and size are similar to those of fulvic acids, the mechanism of stabilization is probably the same: an increase in the negative charge of the mineral colloids and a subsequent increase in the electrostatic repulsion forces. The only carbohydrate that had a clear destabilising effect at some of the concentrations tested was alginic acid.

TEM images of inorganic particles still present in different experimental conditions after 310 h are shown at the bottom of Figure 10. In the micrographs, the particles shown are outside the SPC measuring range. Particle concentration is higher after the addition of fulvic acid and lower after adding alginic acid compared to the sample without NOM addition. Although the number of particles present in the TEM grids is too low to allow any statistically significant quantification, the decrease in particle concentrations can be qualitatively explained by the early formation of large aggregates (directly visible with alginic acid).

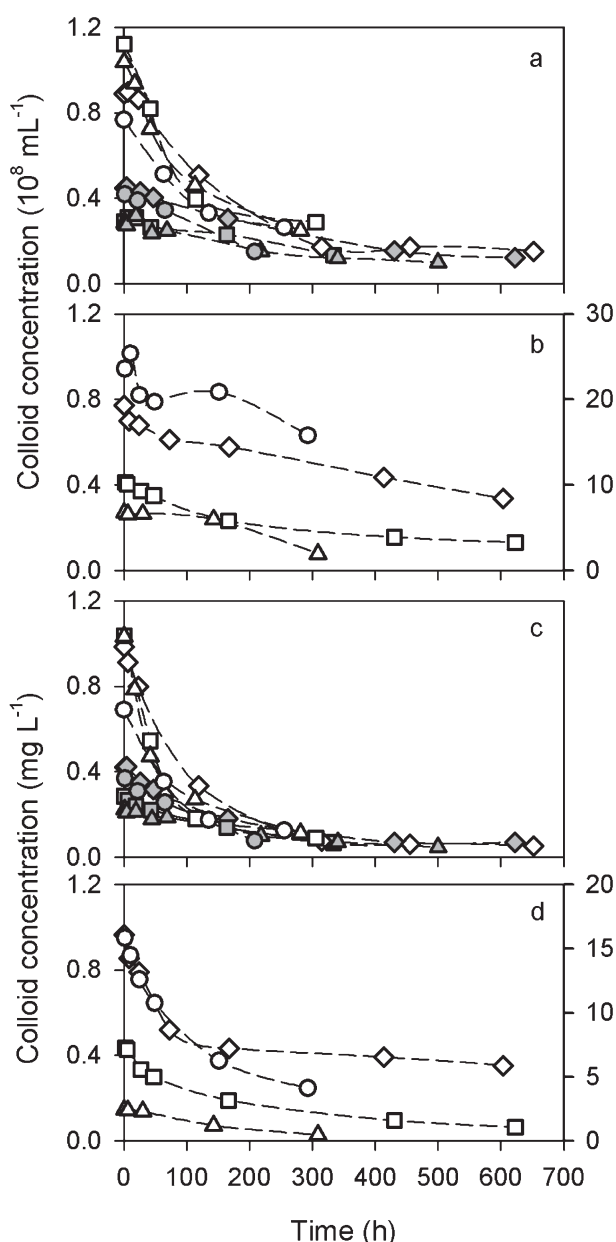


Fig. 9. Variation in number (a) and mass (c) colloid concentrations with time in Lake Brienz waters (1 m depth): \diamond November 2004, \blacksquare February 2005, \blacktriangle March 2005, \bullet April 2005, \diamond July 2005, \square August 2005, \triangle September 2005, \circ October 2005. Variation in number (b) and mass (d) colloid concentrations with time (left axes): \square Lütischine November 2004, \triangle Lütischine February 2005 and right axes: \diamond River Aare November 2004, \circ KWO outlet March 2005.

Discussion

Lake colloid sources

The source of colloids in Lake Brienz is, primarily, its two main tributaries, the Aare and Lütischine. Colloid concentrations in the Lütischine are seasonal dependent. Their large variation is linked to the different production rates of glacial flour from upstream glaciers, which is low in winter and high during the

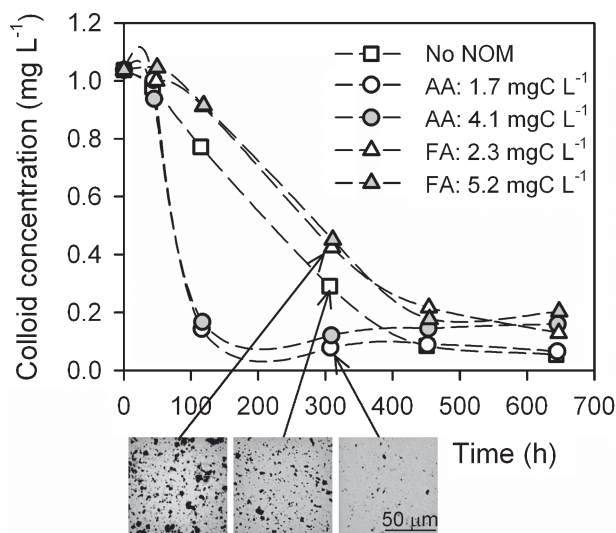
Table 2. Number-coagulation second order rate constants, k_n ; colloid elimination rates, dN/dt ; mass-coagulation second order rate constants, k_m ; temperature; conductivity; DOC and ROM concentrations for the study systems.

System	Date	k_n^a ($10^{-13} \text{ L h}^{-1}$)	$dN/dt \text{ at } t_0^b$ ($10^8 \text{ L}^{-1} \text{ h}^{-1}$)	k_m^a ($\text{L g}^{-1} \text{ h}^{-1}$)	T^c ($^{\circ}\text{C}$)	Conductivity at 25°C^d ($\mu\text{S cm}^{-1}$)	DOC ^e (mg C L^{-1})	ROM ^e (mg C L^{-1})
Lake Brienz, 1 m depth	Nov 2004	2.05 ± 0.08	1.42 ± 0.06	29 ± 2	9.3	167.1	0.35 ± 0.01	0.15^g
	Feb 2005	2.5 ± 0.3	0.74 ± 0.09	34 ± 3	4.8	169.3	0.35 ± 0.01	0.14 ± 0.01
	Mar 2005	2.81 ± 0.09	0.87 ± 0.04	34 ± 2	6.8	169.9	0.68 ± 0.01	0.12 ± 0.01
	Apr 2005	4.4 ± 0.6	2.4 ± 0.3	55 ± 8	8.0	170.7	0.68 ± 0.11	0.18 ± 0.01
	Jul 2005	2.0 ± 0.2	9 ± 1	33 ± 3	15.9	145.0	0.42 ± 0.04	0.11 ± 0.02
	Aug 2005	1.9 ± 0.3	7.5 ± 1.3	36 ± 3	16.1	132.5	nd ^f	nd ^f
	Sep 2005	2.30 ± 0.05	8.2 ± 0.2	30 ± 1	15.2	145.0	0.52 ± 0.04	nd ^f
	Oct 2005	2.0 ± 0.3	6.2 ± 0.8	28 ± 3	15.6	152.9	0.61 ± 0.02	0.15 ± 0.01
Lake Thun	Sep 2005	2.84 ± 0.04	1.20 ± 0.02	46 ± 1	nd ^f	nd ^f	nd ^f	nd ^f
	Oct 2005	3.1 ± 0.2	0.38 ± 0.02	62 ± 7	nd ^f	nd ^f	nd ^f	nd ^f
River Aare	Nov 2004	0.017 ± 0.001	25 ± 1	0.5 ± 0.1	1.3	114.2	0.30 ± 0.09	0.10^g
River Lütschine	Nov 2004	1.75 ± 0.03	1.08 ± 0.03	21 ± 1	2.5	238.0	0.67 ± 0.05	0.13^g
	Feb 2005	1.7 ± 0.2	0.20 ± 0.02	51 ± 5	3.1	337.0	0.77 ± 0.01	0.19 ± 0.04
KWO outlet	Mar 2005	0.017 ± 0.003	34 ± 6	0.61 ± 0.03	2.3	29.5	0.47 ± 0.14	0.26 ± 0.11

^a Error: 1 SD.^b Calculated between 0.1 and 2 μm . Error: 1 SD.^c Error associated with temperature measurements: $\pm 0.2^{\circ}\text{C}$.^d Error associated with conductivity measurements: 1.5 %.^e Chanudet and Filella, 2007. Error: 1 SD.^f nd = not determined.^g For technical reasons, no replicates were made.**Table 3.** Ratio between rate constants measured in the presence of model organic compounds (k_{product}) and rate constants measured in Lake Brienz water (k_{ref}).

Organic substances		Concentration (mg C L^{-1})	$k_{\text{product}} / k_{\text{ref}}$
Carbohydrates	Alginic acid	0.3	0.9
		1.7	2.7 ± 0.5
		4.1	2.0
	Dextran	1.7	1.1
		1.7	0.4
IHSS fulvic acid	Polygalacturonic acid	1.7	1.2
		2.3	0.8
		5.2	0.8

melt season in summer. Glacier colloid production is also high during periods of high river flow (summer), and low glacial colloid production coincides with low flows (winter). As a consequence, there is a large difference between colloid loads to Lake Brienz from the Lütschine in summer and winter. For instance, only 2 tons were input during January 2005 compared to 900 tons in July 2005. The situation is different in the Aare because river flows have been significantly modified by dams and the main source of particles is upstream reservoirs where the two main glaciers drain. Colloid concentrations in these reservoirs are very high (e.g. a colloid concentration of 35 mg L^{-1} was measured at a depth of 1 m in the main reservoir, Grimsensee, in July 2005) and remain relatively constant throughout the year, including winter. The

**Fig. 10.** Variation in mass colloid concentrations with time at different conditions: natural Lake Brienz water (1 m depth) and the same water in the presence of two different concentrations of alginic acid (AA): 1.7 and 4.1 mg C L^{-1} and two different concentrations of fulvic acid (FA): 2.3 and 5.2 mg C L^{-1} . TEM micrographs of particles present in different experimental conditions after 310 h (without the addition of natural organic matter, with the addition of 2.3 mg C L^{-1} fulvic acid and of 1.7 mg C L^{-1} of alginic acid) are shown at the bottom of the figure.

main effect is that the natural seasonal variation in colloid concentrations in unmodified glacial rivers are greatly reduced in the Aare, having much higher winter concentrations than in the Lütschine. On the other hand, Aare flows are dependent on the oper-

ation of the hydroelectrical power stations upstream, resulting in higher discharges in winter and lower discharges in summer compared to the Lütischine and to the Aare's unperturbed state prior to dams (Finger et al, 2006; Anselmetti et al., 2007). As a consequence, the seasonal variation in colloid load differs significantly between the rivers. Although lower colloid loads occur in winter (365 tons during January 2005) than in summer (1130 tons during July 2005) in the Aare, the difference between summer and winter loads is two orders of magnitude less than in the unperturbed Lütischine.

The fate of colloids in the lake

Turbidity measurements provide insight into how colloids are transported from rivers to the lake epilimnion. They show that the Aare and Lütischine enter Lake Brienz as plunging inflows. This is expected because tributary inflows in Lake Brienz have a higher density compared to lake surface waters because of the lower temperature and higher suspended load. The waters of the Aare reach neutral buoyancy in Lake Brienz just above the thermocline in summer, which causes the observed turbid plume. For the Lütischine, a slightly higher water density (higher particle and ion concentration) leads to a slightly deeper intrusion in summer, at the level of the thermocline (Finger et al., 2006). The presence of a turbid plume at the base of the epilimnion and of an upwelling transport from this plume is an important source of epilimnion colloids. An estimate of upwelling speed, based on river inflow values combined with lake geometry, gives upwelling values at a depth of 20 m of 40–50 cm d⁻¹ in summer and 5–10 cm d⁻¹ in winter. These values correspond to Stokes' settling velocities for particles with diameters of about 2.5 µm in summer and 1 µm in winter (particle density = 2.65 g mL⁻¹). Both rivers plunge to the bottom of the lake in winter. A direct result is that much lower colloid concentrations are present in winter in lake surface waters, as confirmed by SPC measurements.

The results from simple one-box mass balance calculations on a whole lake scale show that between June 2004 and July 2005 about 90 % of the colloids in the measured size range were 'lost' through the formation of aggregates > 2 µm, and so outside of the SPC measurement range. A high proportion of these aggregates should sediment to the lake bottom. However, a significant number of particles, outside the colloidal size range, remain in suspension in the surface waters of Lake Brienz. A comparison of mean colloid and particles masses shows that about 80 % of the suspended particles are > 2 µm (Table 1). The high proportion of suspended particles present at a depth of 1 m in Lake Brienz is surprising because a

lake is usually considered an ideal settling basin. The combined effect of two factors can explain these observations. On the one hand, the values given for the maximum diameters expected for upwelling particles are conservative estimates because, in summer, epilimnion mixing mechanisms probably bring particles from the turbid plume to the lake surface that are larger than the estimate based on upwelling considerations. On the other hand, the shape and surface characteristics of the particles (almost exclusively aluminosilicate thin sheets) do not favour sedimentation. Centrifugation results confirmed that Lake Brienz colloids and particles sediment much more slowly than Stokes' law predicts, and that the deviation becomes even more pronounced as the size of the colloids and particles increases.

Insights gained through laboratory measurements

Lake Brienz colloids coagulate relatively slowly in quiescent conditions in the laboratory. Colloid coagulation depends on the mineralogical composition of the colloids and the water chemistry. In the systems studied, the colloid mineralogical composition is similar. As mentioned, the majority of colloids present in the lake are phyllosilicates, in particular 2:1 minerals (micas and illite) (about 60%) and to a lesser extent 2:1+1 minerals such as chlorite (about 10%). Other minerals include Na and K-feldspars (about 30%) and quartz (< 5%). Oxides are almost absent (< 1%) (Filella and Chanudet, 2006). Since most of these colloidal particles show a net negative charge at pH 8, particle aggregation is likely to be severely hindered by surface electrostatic energy. Electrophoretic mobility measurements taken in lake and river samples (results not shown) confirmed the net negative charge of the particle surfaces.

Conductivity values for the Lütischine are higher than for Aare and KWO outlet waters (Table 1) because of its partially calcareous bedrock. The higher ionic concentration explains the higher coagulation rates observed in the Lütischine compared to the Aare and KWO outlet waters for the same sampling campaign (November 2004) (Ali et al., 1984; Edzwald et al., 1974; Weilenmann et al., 1989).

Colloid elimination by coagulation in a given system depends not only on the value of the kinetic constant associated with the type of particles in the water, but also on the number of particles present. Since coagulation is second-order with respect to particle concentration, for any given value of a rate constant, the decrease in particle number will be faster in highly turbid systems, like in Lake Brienz, than in clearer systems. Values for colloid 'effective' elimination rates are shown in Table 2. For instance, for an equivalent time period, about 10 × more colloids are

eliminated in Lake Brienz than in Lake Thun in September and October. In practice, coagulation works as an internal regulation mechanism governing the number of colloids present in a given system and leads to 'pseudo' steady-state conditions. In February, when the colloid concentration in the lake was very low, 0.4 % of the colloids are eliminated in 1 h, but in August and September, when the colloid concentration at the surface was at its highest, about 1 % of the colloids are eliminated in 1 h ($2.5 \times$ faster). This effect is well-known from other systems; e.g. the differing impact of coagulation in different zones of the ocean (i.e., nepheloid layer versus mid-depth regions) and has been attributed to the different number of particles present (McCave, 1975).

Laboratory mass-based coagulation rate constants, k_m , are much higher than the 'effective' lake elimination rate, k , calculated above from mass-balance considerations. While k gives average colloid half life values of 43 days, a half time from 2 to 8 days can be calculated from k_m values depending on the campaign. These differences stem from the different meanings of both constants. For instance, k_m describes coagulation processes under ideal quiescent conditions, is independent of colloid concentrations, and only reflects the coagulation properties of Lake Brienz particles in actual Lake Brienz water chemistry. The 'effective' lake elimination rate constant, k , however, embraces colloid coagulation, the effect of complex lake hydrodynamics (non-quiescent conditions) and the continuous colloid input in the epilimnion by lake transport processes such as upwelling from the turbid plume in summer. The net effect of all processes other than strict colloid coagulation is to increase the 'effective' colloid half life in lake surface waters.

Colloid coagulation and lake productivity

Very slight seasonal variations were observed in the amount and type of NOM present in the lake samples (Table 2). NOM concentrations are very low and relatively constant throughout the year in Lake Brienz (Chanudet and Filella, 2007). No significant change in the coagulation rate during the year due to the presence of NOM would therefore be expected. Nevertheless, the two highest experimental coagulation rates were found in March and April 2005, when carbohydrate concentrations were also highest, being produced by algal and bacteria during the spring bloom. In contrast to humic-type substances (Ali et al., 1984; Jekel, 1986; Amal et al., 1992), some types of carbohydrates have been suggested to favour inorganic colloid coagulation rates (Buffle et al., 1998). The contrasting effects that fulvic acids and carbohydrates are expected to have on the coagulation of lake colloids were confirmed in laboratory experiments for

Lake Brienz colloids. The results showed that a six-fold increase (0.3 to 1.7 mg C L^{-1}) in the carbohydrate concentration would lead, in the best case scenario (alginic acid), to a three fold increase in the coagulation rate. The results support the idea that an increase in primary production in Lake Brienz would lead to an increase in the concentration of carbohydrates present, with a subsequent increase in colloid coagulation rates and decrease in lake turbidity. The increase in surface lake clarity would in turn have a positive effect on lake productivity and particle coagulation rates. However, given the difficulty involved in extrapolating laboratory results to field conditions, as well as the non-linear effect that an increase in coagulation rate constants would have on lake colloid concentrations, it is not possible to quantify the extent of such positive feedback.

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